

Study on the Effect of AO-Coupled Constructed Wetlands on Conventional and Trace Organic Pollutant Treatment

Qianyi Cai, Jingwen Zeng, Xiaojun Lin, Di Xia, Weida Yu, Jinrong Qiu, Mei Yang*,§
and Xiujuan Wang*,§



Cite This: *ACS Omega* 2023, 8, 38983–38990



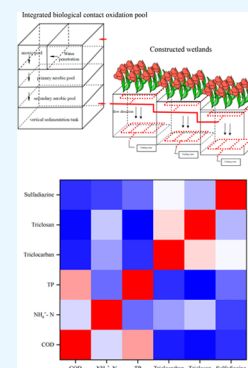
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ABSTRACT: In this study, a pilot-scale integrated process was developed, which combined the integrated biological contact oxidation technology (AO) and the improved constructed wetland technology. The results showed significant removal efficiency for both conventional and trace organic pollutants. The average removal efficiencies for COD, $\text{NH}_4^+\text{-N}$, and TP were 78.52, 85.95, and 49.47%, respectively. For trace organic pollutants, triclocarban, triclosan, and sulfadiazine, the removal efficiencies reached 60.14, 57.42, and 84.29%, respectively. The AO stage played a crucial role in removing trace organic pollutants, achieving removal efficiencies of 37.28, 43.44, and 83.82% for triclocarban, triclosan, and sulfadiazine, respectively. Subsequent treatment using improved constructed wetland technology with coal slag + gravel fillers demonstrated the highest removal efficiency, with average efficiencies of 68.66, 63.38, and 81.32% for triclocarban, triclosan, and sulfadiazine, respectively. Correlation analysis revealed positive correlations between temperature, precipitation, and the removal efficiency of COD, $\text{NH}_4^+\text{-N}$, and TP, while negative correlations were observed with the removal efficiency of triclocarban, triclosan, and sulfadiazine. Furthermore, the influent concentrations of triclocarban and triclosan were significantly negatively correlated with the removal efficiency of COD and TP. The presence of triclocarban and triclosan potentially reduced the microbial diversity and hindered sludge sedimentation performance.



1. INTRODUCTION

In 1999, Daughton and Ternes first introduced the concept of pharmaceuticals and personal care products (PPCPs), drawing international focus onto these substances.¹ These PPCPs, comprising a broad spectrum of substances such as human and veterinary drugs, additives, and inert ingredients in consumer chemical products along with those involved in their production and processing, represent an extensive category of trace organic pollutants.²

Triclosan and triclocarban are prominent members of this group, widely employed for their antibacterial properties in numerous personal care products.^{3–5} In parallel, sulfadiazine, an antibiotic commonly used within the domains of medicine and aquaculture, also falls within this classification.⁶ Consequently, this study focused on triclocarban, triclosan, and sulfadiazine as representative PPCPs.

Despite their comparatively low concentrations in the environment, these trace organic pollutants—triclosan, triclocarban, and sulfadiazine—are ubiquitously detected in varied environmental matrices, such as surface water,⁷ groundwater,⁸ and soil.⁹ The potential genetic toxicity of these pollutants could pose long-term risks to human health.^{10–12} The conventional wastewater treatment processes, not specifically designed to eliminate these trace organic pollutants, inadvertently contribute to their prevalence as the effluent discharge from these facilities constitutes a significant source of such pollutants.^{13,14}

Consequently, these pollutants may enter natural water bodies through inadequately treated wastewater, surface runoff, and landfill leachate seepage.

With the rapid pace of urbanization and socioeconomic development, the focus of wastewater treatment processes has evolved over time.¹⁵ Now, the control indices for wastewater treatment not only incorporate conventional parameters such as COD and $\text{NH}_4^+\text{-N}$ but also extend to trace organic pollution indices. While existing research mainly focuses on individual studies related to the removal of specific types of trace organic pollutants, the unique characteristics of PPCPs and their widespread presence in water bodies necessitate a more comprehensive investigation of their complex interactions and mutual influences.

Wastewater treatment approaches have transitioned from employing single traditional methods to adopting combinations of multiple techniques. A typical approach in wastewater treatment integrates biological contact oxidation with constructed wetlands. Owing to its multiple benefits such as high

Received: May 18, 2023

Accepted: October 4, 2023

Published: October 16, 2023



treatment efficiency, minimal spatial footprint, and strong resistance to load impact, the integrated biological contact oxidation technology has found widespread application in practical sewage treatment. Abbass Jafari Kang et al.¹⁶ explored the removal efficiency of the antibiotic sulfamethoxazole in anoxic/anaerobic/oxic sequencing batch reactors, where they introduced either suspended or granular activated sludge. The granular and suspended biomass achieved sulfamethoxazole removal efficiencies of 84 and 73%, respectively, after 12 h of hydraulic retention time (HRT).

Constructed wetlands, a type of ecological treatment technology, have gained prominence in the advanced treatment process of sewage treatment plants for their low operating costs, ease of maintenance, and environment-friendly characteristics, not to mention their integration with river landscapes.¹⁷ In 2017, Bo Yang¹⁸ formulated a removal model for sulfamethoxazole in vertical flow wetlands, which significantly improved the quantitative balance process in a wetland system. Mark Button et al.,¹⁹ in 2019, utilized a plastic bucket to simulate an enhanced vertical subsurface flow constructed wetland for the removal of triclosan and sulfamethoxazole, both of which exhibited high removal efficiency (>80%). Riyu Zhou et al.,²⁰ in 2022, established a vertical flow constructed wetland, introducing fillers like clay, zeolite, and canna to treat conventional pollutants. Under outdoor dynamic conditions, the average removal efficiencies were found to be 84.38% for COD, 65.78% for $\text{NH}_4^+\text{-N}$, and 74.67% for TP. However, the investigation into the removal of various trace organic pollutants is still largely experimental and lacks comprehensive studies, thus emphasizing the pressing need to develop treatment technologies prioritizing water quality and safety.

This study is centered around the treatment of domestic sewage that contains both conventional pollutants, such as nitrogen and phosphorus, and trace organic pollutants, including triclosan, triclocarban, and sulfasalazine. To tackle the water quality risks associated with these pollutants, researchers developed a pilot test device that integrates biological contact oxidation technology (AO) with improved constructed wetland technology. The aim is to examine the removal efficiency of trace organic pollutants using a combined process of biological contact oxidation and constructed wetland, assess the effectiveness of different fillers on trace organic pollutants, identify the most efficient fillers, and probe the factors influencing the removal process. The ultimate objective is to establish groundwork for the engineering application of advanced treatment technology for this type of wastewater, thereby contributing to the enhancement of water quality and the protection of the environment.

2. TEST EQUIPMENT AND METHOD

2.1. Design Water Intake. The pilot-scale integrated process is situated near Chebei Yong in Guangzhou. The location is shown in Figure 1. The Chebei Yong begins from the Longdong Reservoir and drains into the Pearl River's front channel at Dongpu, spanning a total length of 19.40 km and covering a catchment area of 73.65 km².²¹

The experimental design uses tap water to simulate water intake, which is then supplemented with ammonium sulfate, potassium dihydrogen phosphate, glucose, triclosan, and sulfonamide antimicrobials. The specific concentrations of these components are listed in Table 1.

2.2. Process Route. A pilot study was conducted to develop and evaluate a combined process of AO integrated biological

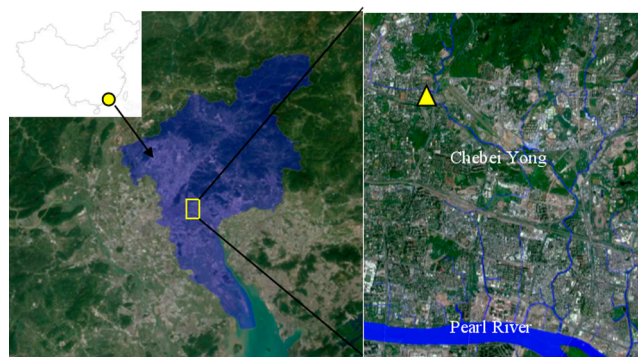


Figure 1. Location of the pilot test device of the integrated process.

Table 1. Inlet Water Quality of the Experimental Device

| COD (mg·L ⁻¹) | $\text{NH}_4^+\text{-N}$ (mg·L ⁻¹) | TP (mg·L ⁻¹) | triclocarban (mg·L ⁻¹) | triclosan (mg·L ⁻¹) | sulfadiazine (mg·L ⁻¹) |
|------------------------------|---|-----------------------------|---------------------------------------|------------------------------------|---------------------------------------|
| 80–120 | 20–40 | 2–4 | 0.1–0.5 | 0.1–0.5 | 0.1–0.5 |

contact oxidation technology and improved constructed wetland technology, with a designed water volume of 10 m³/d. The process is shown in Figure 2.

2.3. Design and Parameters of the Main Structures.

The integrated biological contact oxidation technology (AO) has dimensions of 2.6 m × 1 m × 3.5 m. It is compartmentalized into several sections: an anoxic pool, a primary aerobic pool, a secondary aerobic pool, and a vertical sedimentation tank. The wastewater first enters the anaerobic tank for anaerobic acidification and is then transferred in fixed intervals and volumes to the anoxic tank for further treatment. Following this, the wastewater is directed to primary and secondary oxidation tanks for aeration treatment. Post-treatment, the wastewater enters the secondary sedimentation tank for settling and is subsequently discharged. The anoxic pool, primary aerobic pool, secondary aerobic pool, and vertical sedimentation tank each have an effective volume of 0.8 × 3.0 × 3.5, 0.6 × 3.0 × 3.5, 0.6 × 3.0 × 3.5, and 0.6 m × 3.0 m × 3.5 m, respectively. The HRT stands at 15 h, with an air–water ratio of 4:1.

The improved constructed wetland technology is positioned adjacent to the integrated equipment, providing secondary treatment for the pretreated wastewater from the integrated process. Local canna varieties were chosen as experimental plants for the constructed wetlands, given their high nutrient absorption capability, adaptability to local climate conditions, robust growth, ease of disposal and use, and aesthetic ecological landscape value. To enhance the treatment performance of the constructed wetlands, a variety of common functional fillers with better adsorption capacity and lower price were selected, including coal slag, ceramsite, and gravel, combinations of coal slag and gravel, ceramsite and gravel, and zeolite and gravel. These fillers were deployed across six groups of vertically flow constructed wetland treatment units. The relevant parameters of the constructed wetland fillings can be found in Table 2. The constructed wetlands were 1 × 1 × 1 m cement ponds, filled to a depth of 0.8 m. In the combined constructed wetlands, each type of filler was applied to a depth of 0.4 m. The wetland plants used were local canna varieties, planted at a density of 9 trees/m².

2.4. Analysis Method. During the experimental stage, routine water sample collection was performed from the catchment basin's effluent, the integrated effluent, and the effluent from the six groups of constructed wetlands. Each

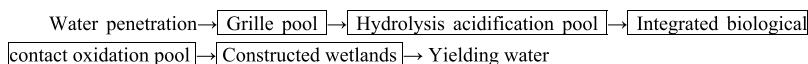


Figure 2. Domestic sewage treatment process.

Table 2. Related Parameters of Constructed Wetland Fillings

| handle name | filling and its thickness | filler particle size |
|--------------------|--|-------------------------------|
| coal slag | 0.8 m cinder | 10–30 mm coal slag |
| ceramsite | 0.8 m ceramsite | 10–30 mm ceramsite |
| gravel | 0.8 m gravel | 10–30 mm gravel |
| coal slag + gravel | 0.2 m coal slag (upper layer) + 0.6 m gravel (lower layer) | 10–30 mm coal slag and gravel |
| ceramsite + gravel | 0.2 m ceramsite (upper layer) + 0.6 m gravel (lower layer) | 10–30 mm ceramsite and gravel |
| zeolite + gravel | 0.2 m zeolite (upper layer) + 0.6 m gravel (lower layer) | 10–30 mm zeolite and gravel |

sampling event, conducted a minimum of six times a month, involved the collection of eight water samples.

Chemical oxygen demand (COD) was determined using the potassium dichromate method, ammonium nitrogen ($\text{NH}_4^+\text{-N}$) was analyzed using Nessler's reagent spectrophotometry, and total phosphorus (TP) was quantified using molybdenum antimony spectrophotometry. Trace organic pollutants were measured as follows: triclocarban and triclosan were analyzed by using liquid chromatography-tandem mass spectrometry (LC-MS/MS), while sulfadiazine was quantified by using high-performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS).

2.4.1. LC-MS/MS Condition. Chromatographic separation of extracts used an Agilent 1290 Infinity HPLC (Agilent Corporation, CA, USA) equipped with a Online solid phase extraction column C18 with a reversed-phase analytical column (Purospher STAR RP-18e, 4.6 mm \times 250 mm, 5 μm particle size; Merck, Germany) held at 30 $^\circ\text{C}$. Binary mobile phases consisting of DI water (A) and methanol (B) were used, with a flow rate of 0.6 mL/min.²²

Analyte detection used a triple-quadrupole mass spectrometer (Agilent G6420) with electrospray ionization (ESI+/ESI– mode). Nitrogen was used for both nebulizing and desolvation gases (300 $^\circ\text{C}$, 10 L/min flow rate). Capillary voltages were 3000 V for both the ESI+ and ESI– modes. In the multiple reaction monitoring (MRM) mode, two individual ion transitions (where possible) were monitored. The ion with a higher peak area response was used for quantification, and the second was used for qualification and identity confirmation. MS/MS parameters including precursor and product ions and their ratios, fragmentor voltage, and collision energy for all target analytes and isotope-labeled internal standards were optimized using Agilent MassHunter Optimizer software.

2.4.2. HPLC-MS/MS Conditions. Chromatographic separation of extracts used an Agilent 1290 Infinity HPLC (Agilent Corporation, CA, USA) equipped with a Online solid phase extraction column C18 with a reversed-phase analytical column (Purospher STAR RP-18e, 4.6 mm \times 250 mm, 5 μm particle size; Merck, Germany) held at 30 $^\circ\text{C}$. Binary mobile phases consisting of acetonitrile (A) and 0.1% formic acid–5 mM ammonium formate aqueous solution (B) were used. The gradient program was set as follows: started at 20% A, 40% A at 8 min, 70% A at 11 min, 80% A at 13 min, 80% A at 15 min, and 20% A at 16 min, with a flow rate of 0.6 mL/min.

Analyte detection used a triple-quadrupole mass spectrometer (Agilent G6420) with electrospray ionization (ESI+/ESI– mode). Nitrogen was used for both nebulizing and desolvation gases (325 $^\circ\text{C}$, 10 L/min flow rate). Capillary voltages were 4000 V for both the ESI+ and ESI– modes. In the MRM mode, two individual ion transitions (where possible) were monitored. The ion with a higher peak area response was used for quantification and the second for qualification and identity confirmation. MS/MS parameters including precursor and product ions and their ratios, fragmentor voltage, and collision energy for all target analytes and isotope-labeled internal standards were optimized using Agilent MassHunter Optimizer software.

2.4.3. Analytical Method Validation. The recoveries of the three target compounds were determined by using the standard addition method. Calibration curves were constructed by plotting the ratio of the analyte quantifying the ion response to the internal standard (IS) ion response against the spiked concentrations. The correlation coefficients (r^2) for all the antibiotics exceeded 0.99, indicating excellent linearity within the selected concentration range. The limits of detection and limits of quantification, defined as the lowest concentration levels corresponding to signal-to-noise ratios of 3 and 10, respectively, were also determined.

3. RESULTS AND ANALYSIS

3.1. Study on the Pollutant Removal Effect. A comprehensive long-term monitoring program was implemented to assess the quality of the effluent from the integrated inlet and outlet water as well as from the six groups of constructed wetlands. The study involved modifying the volume of aeration in the integrated system and analyzing the resultant average removal efficiency of conventional and trace organic pollutants for each treatment unit. Further, a comparative analysis was conducted to ascertain the average removal efficiency of pollutants before and after the aeration adjustments.

In tandem with this, six groups of constructed wetlands were established, utilizing functional fillers such as coal slag, ceramsite, and gravel, a combination of coal slag and gravel, a mixture of ceramsite and gravel, and zeolite combined with gravel. The primary objective of this aspect of the study was to ascertain the efficacy of these diverse functional fillers in the mitigation of both conventional and trace organic pollutants within the confines of the constructed wetlands. Through rigorous and extensive experimental research, the fillers that contributed the most significantly to enhancing the functionality of the wetlands were identified. Ultimately, the study aimed to establish optimal practices for the effective utilization of constructed wetlands in the treatment of water contaminated with a range of pollutants.

3.1.1. Conventional Pollutants. Figure 3 illustrates that the COD removal efficiency in sewage post integrated treatment is 61%, while after undergoing intensive treatment in a constructed wetland, it can reach between 77 and 82%. The average COD effluent concentration was found to be within 29.14 to 42.26 mg/L. COD removal in constructed wetlands is primarily accomplished through microbial aerobic and anaerobic

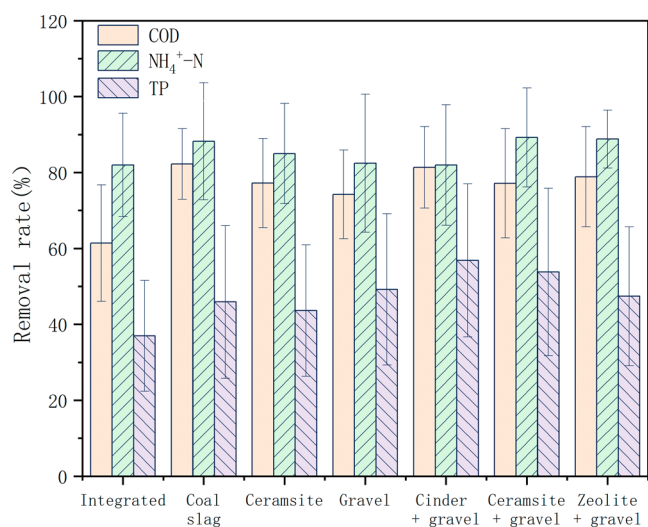


Figure 3. Removal effect of each treatment unit on conventional pollutants.

degradation, filler adsorption, sedimentation, and plant absorption. The majority of organic matter in domestic sewage is adsorbed by the wetland filler and its surface biofilm and is then gradually degraded by microorganisms, thus enhancing the COD removal efficiency.²³ The most effective COD removal was observed when coal slag was used as a wetland filler. This can be attributed to the numerous pores, large specific surface area, and good air permeability of coal slag, which facilitate the microbial aerobic degradation of organic matter.

The NH₄⁺-N removal efficiency in sewage post integrated treatment is 82%, and the average removal efficiency can hit between 82 and 89% after intensive treatment in a constructed wetland. The average NH₄⁺-N effluent concentration was observed to be within 3.10 to 5.13 mg/L. The integrated system is highly effective at NH₄⁺-N removal, which is primarily a process of microbial degradation and plant absorption. Wetland fillers provide a favorable environment for microorganisms and plants.²⁴ The best NH₄⁺-N removal results were achieved when ceramsite + gravel was used as the wetland filler.

The TP removal efficiency in sewage post integrated treatment is 40%, while the average TP removal efficiency after enhanced treatment in the constructed wetland ranges from 46 to 57%. The average TP effluent concentration was observed to be within 3.29 to 5.29 mg/L. The most effective TP removal was achieved when coal slag + gravel was used as a filler, which might be attributed to the good air permeability of the gravel, loose and porous coal slag, and the presence of iron, aluminum, and their oxides, as well as calcium compounds that enhance the adsorption of phosphorus by the wetland filler. Compared to COD and NH₄⁺-N removal efficiencies, TP removal efficiency is relatively lower. TP is primarily removed through chemical precipitation, microbial metabolism, and plant absorption, but the efficacy of microbial phosphorus removal is subpar. Chemical precipitation with metal ions such as Fe³⁺, Ca²⁺, and Al³⁺ present in the sewage is the principal mode of phosphorus removal. The increase in the TP removal efficiency of each treatment unit in the latter stage might be due to the growth of constructed wetland plants.²⁵

As depicted in Figure 4, alterations in the aeration efficiency significantly impact the removal of COD and TP, with the removal efficiency rising as the aeration efficiency increases. Within a certain range, aeration efficiency and dissolved oxygen contribute positively to the removal of COD. As with TP, it is mainly removed by chemical precipitation, microbial metabolism, and plant absorption, among which chemical precipitation with metal ions such as Fe³⁺, Ca²⁺, Al³⁺, and other ions present in sewage is the primary mode of phosphorus removal. An increase in the aeration efficiency can encourage the coprecipitation of metal ions and phosphorus. However, changes in the aeration efficiency appear to have a minimal effect on the NH₄⁺-N removal efficacy.

3.1.2. Trace Organic Pollutants. Figure 5 shows that the removal efficiencies of triclocarban and triclosan after the integrated treatment can reach 37 and 43%, respectively. After undergoing treatment in constructed wetlands, these removal efficiencies can increase to ranges of 54 to 69 and 51 to 64%, respectively. The average effluent concentrations of triclocarban and triclosan were measured to be within 8.24 to 13.17 and 12.78 to 21.90 ng/L, respectively.

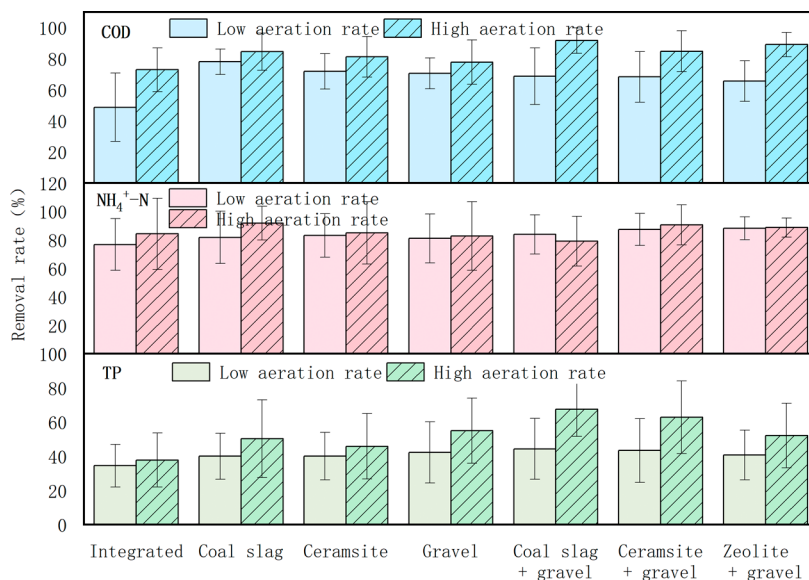


Figure 4. Removal efficiency of conventional pollutants under different aeration efficiencies.

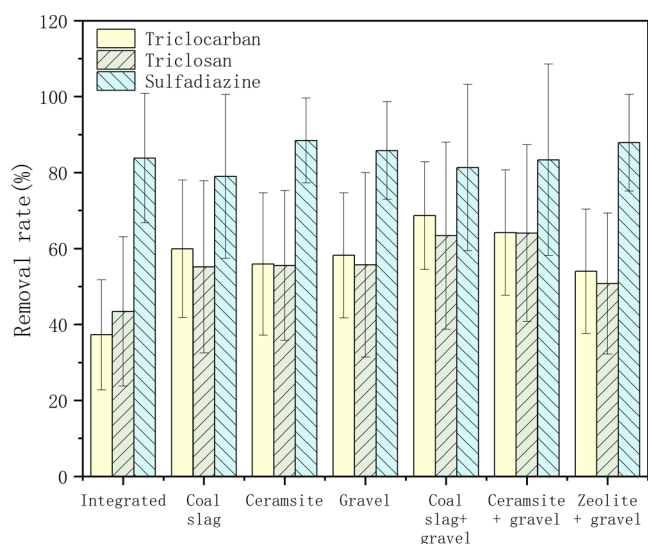


Figure 5. Removal effect of each treatment unit on trace organic pollutants.

Meanwhile, the removal efficiency of sulfadiazine after integrated treatment can reach 84%, and after treatment in constructed wetlands, it can increase to a range of 79 to 88%. The average effluent concentration of sulfadiazine was measured to be between 0.94 and 1.71 ng/L. Constructed wetlands appear to enhance the removal effect of sulfadiazine. However, the effluent concentration of sulfadiazine in the constructed wetland treatments using (ceramsite + gravel), (coal slag + gravel), and (coal slag) as fillers was higher than that of the integrated treatment. This could be due to the adsorption of sulfadiazine in the wetland fillers and its subsequent release into the water through desorption, resulting in increased effluent concentrations.

As illustrated in Figure 6, changes in the aeration efficiency significantly impact the removal of triclocarban and triclosan, with the removal efficiency of these two pollutants decreasing as the aeration efficiency increases. The change in the aeration efficiency seems to have a minor effect on the removal efficiency

of sulfadiazine. The removal efficiency of the hydrophobic compounds triclocarban and triclosan tends to decrease with increasing aeration efficiency, suggesting that adsorption might be the primary removal mechanism.^{26,27} This could be because an increase in aeration disrupts the water body, leading to the desorption of pollutants. Congcong Zhao et al.²⁸ demonstrated the biodegradation of triclosan by constructed wetlands and its mechanism.

3.2. Analysis of Removal Efficiency Correlation.

3.2.1. Analysis of Influencing Factors for Pollutant Removal.

The relationship among influent concentration, precipitation, daily average temperature, and the removal efficiency of each treatment unit was analyzed using SPSS software, with the results displayed in Table 3. The precipitation and daily average temperature data used during the experiment were sourced from the National Oceanic and Atmospheric Administration of the United States. The weather station utilized for data retrieval was situated in Guangzhou, China, with coordinates 23.217N and 113.483E.

Temperature is a vital factor that significantly influences removal efficiency.²⁹ The daily average temperature exhibited a positive correlation with the removal efficiency of COD, $\text{NH}_4^+\text{-N}$, and TP. As the temperature rises, the removal efficiency of COD also improves. This phenomenon can be attributed to higher temperatures promoting plant growth and continuous oxygen release from roots, thus facilitating aerobic degradation processes. Changes in temperature have a minimal impact on the removal of $\text{NH}_4^+\text{-N}$. The mechanisms of removal of ammonia nitrogen in constructed wetlands include $\text{NH}_4^+\text{-N}$ volatilization, plant absorption, packing adsorption, and microbial nitrification and denitrification. In the microbial $\text{NH}_4^+\text{-N}$ removal process, nitrification is a key rate-limiting step where ammonia-oxidizing bacteria and ammonia-oxidizing archaea work synergistically.³⁰ When the temperature drops, the abundance of ammonia-oxidizing bacteria decreases, but ammonia-oxidizing archaea maintain a high abundance, resulting in little impact on the nitrification reaction.³¹ Contrastingly, the daily average temperature was negatively correlated with the removal efficiency of triclosan, triclocarban, and sulfadiazine, with a notably significant negative correlation with triclocarban.

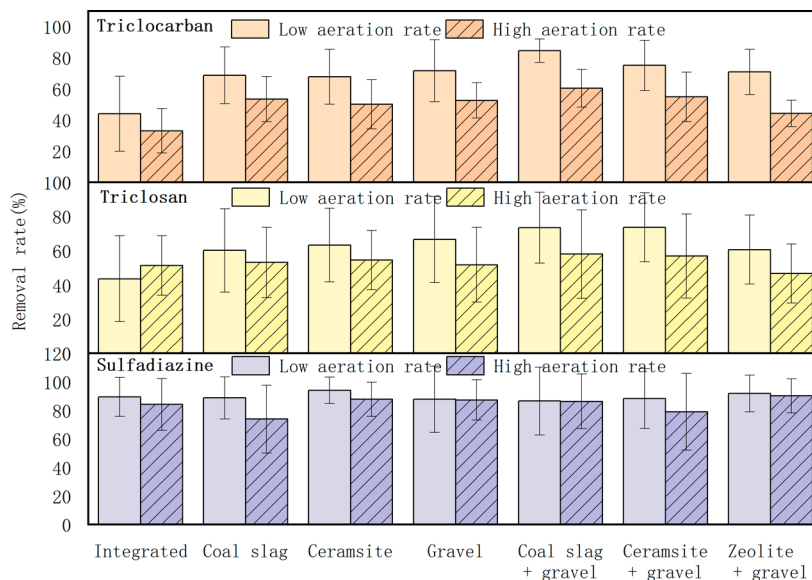


Figure 6. Removal efficiency of trace organic pollutants under different aeration efficiencies.

Table 3. Correlation Analysis of Several Influencing Factors to Pollutant Removal Efficiency^a

| factor processing | removal efficiency | | | | | |
|---------------------------|---------------------------------|---------------------------------|----------|--------------|-----------|--------------|
| | COD | NH ₄ ⁺ -N | TP | triclocarban | triclosan | sulfadiazine |
| daily average temperature | 0.375** | 0.094 | 0.228** | -0.637** | -0.167 | -0.242** |
| precipitation | 0.068 | 0.161* | 0.249** | -0.189* | -0.355** | -0.011 |
| influent concentration | COD | 0.197* | -0.533** | 0.370** | -0.084 | -0.233* |
| | NH ₄ ⁺ -N | 0.168* | -0.215** | 0.341** | 0.008 | -0.243** |
| | TP | 0.461** | -0.291** | 0.751** | -0.515** | -0.526** |
| | triclocarban | -0.275** | -0.009 | -0.281** | 0.538** | 0.277** |
| | triclosan | -0.480** | -0.123 | -0.368** | 0.649** | 0.568** |
| | sulfadiazine | 0.236* | -0.002 | 0.369** | -0.153 | -0.191* |

^a**At the 0.01 level (two-tailed), the correlation is significant. *At the 0.05 level (two-tailed), the correlation is significant.

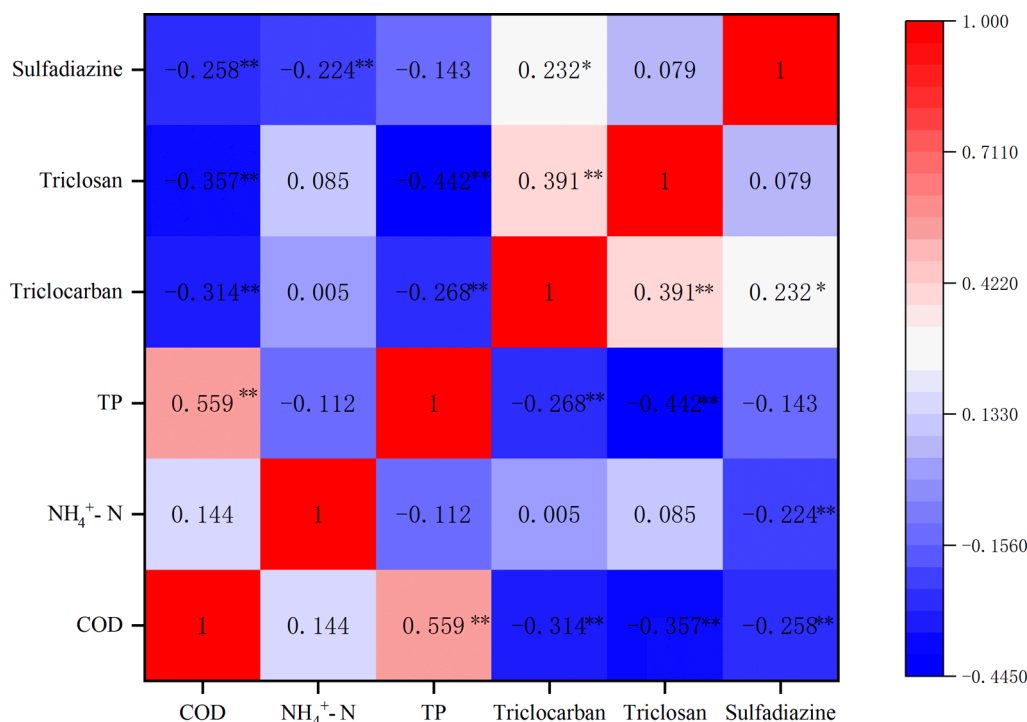


Figure 7. Correlation analysis of the pollutant removal efficiency.

The influent concentrations of COD, TP, triclocarban, and triclosan demonstrated a significant positive correlation with their respective removal efficiencies, with removal efficiency increasing alongside influent concentration. This trend can primarily be attributed to an increased driving force.³² Conversely, the influent concentration of NH₄⁺-N negatively correlated with the removal efficiency of NH₄⁺-N, suggesting that when the adsorption capacity of the wetland filler reaches saturation, the removal efficiency decreases as the influent concentration increases. The influent concentrations of triclocarban and triclosan exhibited a negative correlation with the removal efficiency of COD, NH₄⁺-N, and TP. The presence of triclocarban and triclosan potentially led to a decrease in microbial diversity and interfered with sludge sedimentation in the system, subsequently diminishing the removal efficiency of conventional pollutants.

Precipitation significantly impacts the removal efficiency of NH₄⁺-N, TP, triclocarban, and triclosan. It shows a positive correlation with the removal efficiency of COD, NH₄⁺-N, and TP and a negative correlation with the removal efficiency of triclocarban, triclosan, and sulfadiazine. As precipitation increases, a dilution effect occurs, reducing the concentration

of conventional pollutants in the water body. Therefore, the removal efficiency of COD, NH₄⁺-N, and TP increases, indicating a positive correlation. However, increased precipitation accelerates water flow, impacting the migration of persistent pollutants, such as triclocarban, triclosan, and sulfadiazine. This makes the removal of these recalcitrant pollutants more difficult, hence establishing a negative correlation between precipitation and the removal efficiency of triclocarban, triclosan, and sulfadiazine.

3.2.2. Analysis of the Synergistic Removal Effect of Pollutants. As illustrated in Figure 7, the removal of COD enhances the removal of NH₄⁺-N and TP, showing a significant positive correlation with the removal efficiency of TP. The removal of COD promotes the growth and metabolism of microorganisms, increasing their population and activity. During the process of microbial growth and metabolism, organic matter and extracellular polymers are secreted. These substances have the ability to adsorb and precipitate phosphate in the surrounding environment, facilitating the removal of TP. Additionally, microorganisms can utilize NH₄⁺-N for their growth and metabolism, converting it into inorganic nitrogen, thus achieving the removal of NH₄⁺-N.

The removal of triclocarban exhibited a positive correlation with the removal of triclosan and sulfadiazine, indicating that these pollutants may be influenced by similar environmental factors due to their similar degradation mechanisms and processes in aquatic environments. Triclocarban, triclosan, and sulfadiazine are primarily eliminated through biodegradation in water environments, where organisms decompose these organic compounds into inorganic substances and carbon dioxide through metabolic processes.

The removal efficiency of three trace organic pollutants showed a negative correlation with the removal efficiency of COD, ammonia nitrogen, and TP. This observation may arise from the distinct removal mechanisms and characteristics of these pollutants. Conventional pollutants such as COD, ammonia nitrogen, and TP are primarily eliminated through biodegradation, plant absorption, precipitation, and other mechanisms. However, trace organic pollutants can potentially hinder the growth and metabolism of microorganisms and plants, subsequently diminishing the treatment effectiveness of constructed wetlands. Zhiqi Wang et al. conducted a study indicating that triclosan decreased microbial diversity and altered the microbial community.³³

4. CONCLUSIONS

- 1) A pilot study was conducted to research and develop a combined process that integrates the biological contact oxidation technology (AO) and the improved constructed wetland technology. This combined process exhibited a significant removal effect on simulated domestic sewage. The average removal efficiencies for conventional pollutants, including COD, $\text{NH}_4^+\text{-N}$, and TP, were 78.52, 85.95, and 49.47%. Furthermore, the removal efficiencies for trace organic pollutants, such as triclocarban, triclosan, and sulfadiazine, reached 60.14, 57.42, and 84.29%.
- 2) The combined process of AO integrated biological contact oxidation technology and improved constructed wetland technology demonstrated the effective removal of trace organic pollutants, such as triclocarban, triclosan, and sulfadiazine. The primary removal of these pollutants occurred during the AO stage, with removal efficiencies of 37.28, 43.44, and 83.82% for triclocarban, triclosan, and sulfadiazine, respectively. Following the advanced treatment provided by the improved constructed wetland technology, the performance of six different functional fillers was compared. The constructed wetland using coal slag + gravel as combined fillers exhibited the highest removal efficiency, with average removal efficiencies of 68.66, 63.38, and 81.32% for triclocarban, triclosan, and sulfadiazine, respectively.
- 3) The correlation analysis revealed that the removal efficiencies of COD, $\text{NH}_4^+\text{-N}$, and TP were positively correlated with temperature (daily average air temperature) and precipitation. However, the removal efficiencies of triclocarban, triclosan, and sulfadiazine showed a negative correlation with temperature and precipitation. Furthermore, the influent concentrations of triclocarban and triclosan were significantly negatively correlated with the removal efficiency of COD and TP. The presence of triclocarban and triclosan could potentially reduce microbial diversity and hinder the sedimentation performance of sludge in the system.

AUTHOR INFORMATION

Corresponding Authors

Mei Yang – College of Forestry, Guangxi University, Nanning 530004, China; Email: fjyangmei@126.com

Xiujuan Wang – South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China; Email: 2009592001@st.gxu.edu.cn

Authors

Qianyi Cai – College of Forestry, Guangxi University, Nanning 530004, China; South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China; orcid.org/0009-0005-4650-8437

Jingwen Zeng – South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China

Xiaojun Lin – South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China

Di Xia – South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China

Weida Yu – South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China

Jinrong Qiu – South China Institute of Environmental Sciences, Ministry of Ecology and Environment, Guangzhou 510655, China

Complete contact information is available at:
<https://pubs.acs.org/10.1021/acsomega.3c03461>

Author Contributions

[§]M.Y. and X.W. contributed equally.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was funded by Science and Technology Projects in Guangzhou (202002030276).

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